

5. POTENTIAL FOR HUMAN EXPOSURE

5.1 OVERVIEW

Plutonium is a radioactive element produced by neutron capture and beta decay of uranium-238 (or other elements), both naturally (in minuscule amounts) and as a result of human activities. Plutonium is found in the environment in the form of several isotopes. The source of plutonium can be traced based on the isotope or isotopes detected in a sample. Plutonium is found naturally in uranium-rich ores in concentrations of one part per 10¹¹ parts uranium (i.e., 1×10^{-11} kg plutonium/kg uranium) (Leonard 1980).

The principal plutonium isotopes used in commerce and by the military are plutonium-238 and plutonium-239. These two isotopes are used because of their ease of production and their relatively long half-lives. Plutonium-238 is used in thermoelectric generation systems in spacecraft, cardiac pacemakers, and other power sources (Harley 1980; NEA/OECD 1981). Plutonium-239 and -240 are produced in nuclear power plants as a product of nuclear fission as well as in production facilities for use in nuclear weapons.

Possible sources of plutonium to the environment include: weapons testing, accidents involving weapons transport, nuclear reactors and radioisotope generators, fuel processing and reprocessing, and fuel transport (NEA/OECD 1981). Plutonium-239 is generated in irradiated uranium fuel when neutrons are captured by uranium-238 nuclei. Some of the plutonium-239 is consumed during the operation of the reactor. Production of plutonium by nuclear reactors generating electricity and by weapons production was estimated at 3.8×10^6 kg in 1978 (NEA/OECD 1981).

Atmospheric testing of nuclear weapons has been the main source of plutonium dispersed in the environment. Accidents and routine releases from weapons production facilities are the primary sources of localized contamination. Consumer and medical devices containing plutonium are sealed and are not likely to be environmental sources of plutonium (WHO 1983). Plutonium released to the atmosphere reaches the earth's surface through wet and dry deposition to the soil and surface water. Once in these media, plutonium can sorb to soil and sediment particles or bioaccumulate in terrestrial and aquatic food chains.

According to the NPL database (VIEW 1989), plutonium has been identified above background levels at five NPL sites. Plutonium-238 has been identified at three of these sites, plutonium-239 at five sites, and plutonium-240 at one site. The frequency of these sites within the United States can be seen in Figure 5-1.

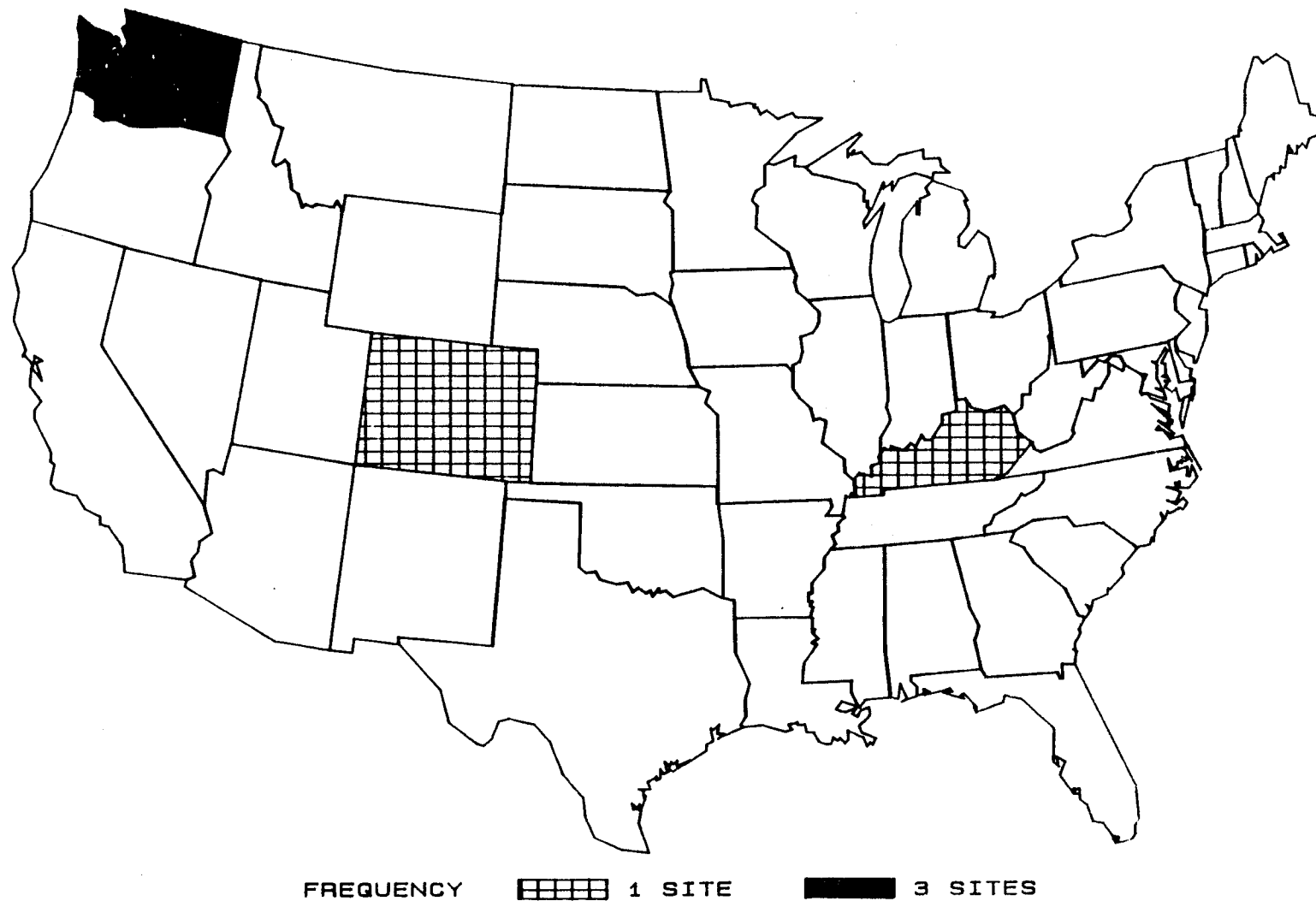


FIGURE 5-1. FREQUENCY OF SITES WITH PLUTONIUM CONTAMINATION

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5.2 RELEASES TO THE ENVIRONMENT

5.2.1 Air

Anthropogenic (man-made) releases of plutonium are the primary sources of plutonium to the atmosphere. Atmospheric testing, fires involving plutonium-containing materials, and routine releases due to normal activities at processing and generating plants are all potential sources of airborne plutonium. Resuspension of plutonium sorbed to contaminated surface soils via fugitive dust emissions is an indirect pathway by which plutonium may be re-released into the atmosphere (Harley 1980).

Plutonium released during nuclear weapons testing is the largest source of plutonium-239 and -240 in the environment (Harley 1980). Approximately 320 kCi (1.2×10^{16} Bq) of plutonium-239, -240 and 9 kCi (3.3×10^{14} Bq) of plutonium-238 have been released to the atmosphere by nuclear tests and distributed worldwide (Eisenbud 1987). Concentrations of transuranics introduced into the environment through underground test venting, accidents involving United States nuclear weapons, and releases during weapon production operations have been negligible in comparison with those released during atmospheric testing of nuclear explosives in the 1960s (Facer 1980).

In April, 1964, a Transit Navigational Satellite was launched in California with a payload that included a Satellite for a Nuclear Auxiliary Power Generator (SNAP-9A) containing 17 kCi (6.3×10^{14} Bq) of plutonium-238. The rocket system failed and the satellite reentered the atmosphere in the Southern Hemisphere and burned over the Indian Ocean at an altitude of about 50 km (Harley 1980). The destruction of the SNAP-9A resulted in the largest single release of plutonium-238 to the atmosphere, primarily in the form of very small oxide particles (Harley 1980).

Research facilities and plants have also released plutonium to the atmosphere. For example, the Mound Plant in Miamisburg, Ohio, released about 0.03 kCi (1×10^{10} Bq) to the atmosphere from the beginning of its operation through 1976 (NEA/OECD 1981). A commercially operated reprocessing plant in West Valley, New York, has reportedly released 0.000005 kCi (1.7×10^8 Bq) to the atmosphere over the course of 6 years (NEA/OECD 1981).

5.2.2 Water

Fallout from atmospheric weapons testing, accidents involving nuclear weapons, planned as well as accidental reactor effluent releases, and disposal of radioactive wastes are all means by which plutonium can be introduced into water systems (Harley 1980; NEA/OECD 1981). In a typical 1,000 megawatt electric (MWe) light water reactor

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in a nuclear power plant about 200 kg of plutonium [equivalent to 13 kCi (4.8×10^{14} Bq); one curie of plutonium-239 - 16g] are generated per year of operation in the spent fuel (NEA/OECD 1981; Facer 1980). Contaminated cooling water containing plutonium from nuclear production facilities may have been discharged into oceans or rivers. If release occurs from waste containers, buried radioactive wastes may migrate or seep into groundwater (NEA/OECD 1981). As an example of plant emissions, the Mound Plant in Miamisburg, Ohio, discharged a total of about 0.0005 kCi (1.9×10^{10} Bq) plutonium-238 into a river near the site from the beginning of its operation through 1976 (NEA/OECD 1981).

In January, 1968, while attempting to make an emergency landing, a United States military aircraft with four nuclear weapons on board crashed in Thule, Greenland. The impact resulted in detonation of the high explosives in all four nuclear weapons aboard. The oxidized plutonium was dispersed by both the explosion and the fire involving the fuel in the jet (Harley 1980). Amounts of plutonium released to the air in this accident have been estimated at 0.024 kCi (9×10^{11} Bq) of insoluble plutonium (NEA/OECD 1981). The maximum concentration of plutonium in ocean sediments was found 1 km from the point of impact. The sediment-bound plutonium was found to migrate both downward in the sediment column and horizontally from the point of impact. The concentrations decreased with distance from the point of impact.

Sediments can act as both a repository for and a source of waterborne plutonium. Atmospheric fallout reaching surface water can settle in the sediments. The plutonium in the ocean sediments at Bikini Atoll, for example, was found to be resuspended and released to the bottom waters (Schell et al. 1980). In a freshwater waste pond at the Hanford reactor, plutonium was found to be bound to the sediments and was not available for uptake by plants or animals in the pond (Emery et al. 1980). The difference between the observations in the two ecosystems may be due to the dynamic nature of the ocean water near Bikini Atoll versus the relatively static nature of a waste water pond.

5.2.3 Soil

Plutonium has been detected in extremely small amounts as a naturally occurring constituent of some minerals and ores. Uranium and thorium ores in Canadian pitchblende, Belgium Congo pitchblende, Colorado pitchblende, Brazilian monazite, and North Carolina monazite have been found to contain plutonium-244 at a weight ratio of up to 9.1×10^{-12} kg plutonium/kg ore (Leonard 1980).

Soils may become contaminated from fallout associated with nuclear weapons tests, such as those conducted at the Trinity Site in southern New Mexico, the Pacific Proving Ground at the Enewetak Atoll, and the Nevada Test Site or with accidental, non-nuclear detonation of nuclear weapons, such as occurred at Palomares, Spain. Research facilities,

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such as the Los Alamos National Laboratory, Los Alamos, New Mexico, may release treated radioactive wastes under controlled conditions. Production facilities, such as the Hanford and Savannah River Plants and experimental reactor stations, for example, the Idaho National Engineering Laboratory, Idaho Falls, Idaho, also released treated plutonium-bearing radioactive wastes under controlled conditions to soils (Hanson 1975).

Atmospheric weapons testing fallout has been a global source of transuranics, including plutonium, in soils (Harley 1980; NEA/OECD 1981). It has been estimated that approximately 100 kCi (3.7×10^{15} Bq) of plutonium from weapons have been distributed globally from all testing sources and could be environmentally available. Of that amount, approximately 1.0 to 10 kCi (3.7×10^{11} to 3.7×10^{12} Bq) were deposited on test site surface soils in the United States (Facer 1980).

Several of the major nuclear facilities in the United States use plutonium and some of these have released plutonium to the environment. These releases have taken place at remote sites and generally have not been measurable outside the plant property. Approximately 0.002 kCi (7.4×10^{10} Bq) of plutonium have been disposed in the Los Alamos National Laboratory canyon waste disposal sites (Harley 1980). The Savannah River Plant, Aiken, South Carolina, has released a total of 0.005 kCi (1.6×10^{11} Bq) of plutonium to local soil (Harley 1980). Leakage of stored waste released between 0.01 and 0.1 kCi (3.7×10^{11} and 3.7×10^{12} Bq) of plutonium to the soil over a period of several years at the Rocky Flats facility, Golden, Colorado (Facer 1980). A break in a waste transfer line caused the release of about 0.3 kCi (1.1×10^{13} Bq) of plutonium-238 at the Mound Plant, Miamisburg, Ohio, in 1969 (Facer 1980).

A fire on May 11, 1969, occurred at the plutonium processing facility at Rocky Flats, Golden, Colorado. Subsequently, a study of the plutonium content in off-site soils was performed. The results of the study indicated that the plutonium found off-site was due, primarily, to small emissions from the facility rather than to the fire, and that a total of 0.003 kCi (9.6×10^{10} Bq) of plutonium was deposited in soils within a 7 mile radius from the facility (Eisenbud 1987).

Another source of soil contamination at Rocky Flats was the leakage of plutonium-contaminated oil. Plutonium was present as the dioxide when it was released. The dioxide was then adsorbed to the soil. Fugitive dust emissions caused plutonium-contaminated soil to be distributed away from the spill. Most of the plutonium remained on the surface, although some was released and migrated downward through the soil column (Little and Whicker 1978).

A United States military aircraft carrying four nuclear bombs collided with a tanker aircraft during refueling in Palomares, Spain, in

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January, 1966. The bombs broke free of the airplane and the high explosive in two of the weapons detonated when the bombs hit the ground. Initial surveys showed that $0.00003 \text{ Ci plutonium/m}^2$ ($1.2 \times 10^5 \text{ Bq/m}^2$), in the form of a finely powdered dioxide, were spread over 2 hectares ($20,000 \text{ m}^2$) (Harley 1980).

5.3 ENVIRONMENTAL FATE

5.3.1 Transport and Partitioning

Plutonium enters the environment primarily through releases to the atmosphere or direct discharge to ponds, streams, or oceans. Emissions to the atmosphere will result in plutonium fallout. In the case of weapons testing, approximately one-fifth of the plutonium released falls on the test site (Harley 1980). The rest is carried in the atmosphere, adsorbed to particulate matter and is transported back to earth via dry or wet deposition. Once plutonium is deposited either on the land or surface water, sorption to soils or sediments is the primary environmental fate of plutonium. A small fraction of plutonium reaching the soil will become solubilized either through chemical or biological processes, depending upon its chemical form. In soluble form, plutonium can either migrate in groundwater or surface water or be available for uptake into plants.

Atmospheric releases of plutonium occur as a result of nuclear weapons testing or routine or nonroutine nuclear reactor operations and fuel reprocessing. The rate at which plutonium is removed from the atmosphere will depend on the chemical and physical properties of particles to which it is adsorbed, as well as the meteorological conditions. The larger the particles, the faster fallout will occur. The particle size expected to be released from either of the above mentioned sources ranges from $0.3 \text{ }\mu\text{m}$ to $1.1 \text{ }\mu\text{m}$. Based on computer modeling, these particles are expected to reach the earth's surface within 60 days of their release (NEA/OECD 1981). The global fallout rate of plutonium-238, predominantly from the SNAP-9 accident, as determined by Harley (1980), is $0.002 \text{ pCi/m}^2/\text{day}$ ($7.4 \times 10^{-5} \text{ Bq/m}^2/\text{day}$) based on plutonium levels measured in surface soils. The global deposition rate of plutonium-239 and plutonium-240 combined is equal to $0.03 \text{ pCi/m}^2/\text{day}$ ($1.1 \times 10^{-3} \text{ Bq/m}^2/\text{day}$) (Corey et al. 1982).

Plutonium deposited on soil surfaces may be resuspended in the atmosphere especially in areas that have low soil moisture levels, such as the Nevada Test Site. In drier areas, the levels of ambient airborne dust are expected to be higher than in areas with normal rainfall (Harley 1980). The highest concentrations of plutonium are likely to be found in the fine silt-clay particle size range. Particles of this size tend to be transported the farthest distance by wind and water (WHO 1983).

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The transport and partitioning of plutonium in soils depends on the form of the compound. The solubility of plutonium depends on the properties of the soil, the presence of organic and inorganic complexing agents, the form of plutonium that enters the soil environment, and the presence of soil microorganisms (Bell and Bates 1988; Kabata-Pendias and Pendias 1984; WHO 1983; Wildung and Garland 1980). Plutonium fallout from the atmosphere, for example, tends to be deposited primarily as the insoluble dioxide (Harley 1980; Wildung et al. 1987). The majority of plutonium remains within the top few centimeters of the soil surface as the dioxide form (WHO 1983). Microorganisms can change the oxidation state of plutonium, thereby either increasing or decreasing its solubility.

Plutonium will migrate in soils as the hydrolyzed ion or as a complex, formed with organic or inorganic acids. Mewhinney et al. (1987b) found that particles subjected to wetting and drying, such as those found on the soil surface, released more plutonium than soils continually immersed in a solvent, such as that found in lakes. This phenomenon is attributed to the formation of a soluble dioxide layer on the particle's surface during the drying phase. Soil organisms have also been found to enhance the solubility of plutonium (Wildung et al. 1987). Once plutonium enters the soluble phase, it then becomes available for uptake by plants. The plutonium(IV) oxidation state is found in plants due to its ability to hydrolyze in the environment (Garland et al. 1981, 1987). Cataldo et al. (1987) postulate that reduction of the higher oxidation states, such as plutonium(VI), occurs prior to absorption/transport across the root membrane.

The behavior of plutonium in surface waters is dependent upon the oxidation state and the nature of the suspended solids and sediments. Plutonium(III) and plutonium(IV) are considered to be the reduced forms of plutonium while plutonium(V) and plutonium(VI) are the oxidized forms. The oxidized forms of plutonium are found in natural waters when the concentrations of dissolved organic matter or dissolved solids are low (Nelson et al. 1987). Humic materials (naturally occurring organic acids) were found to reduce plutonium(V) to plutonium(IV) in sea water. This was followed by adsorption of plutonium(IV) onto iron dioxides and deposition into the sediments (Choppin and Morse 1987).

The partitioning of plutonium from surface water to sediments in freshwater and marine environments depends on the equilibrium between plutonium(IV) and plutonium(V), and the interaction between plutonium(IV) in solution and plutonium sorbed onto sediment particle surfaces (NCRP 1984). Sorption onto marine clays was found to be largely irreversible (Higgo and Rees 1986). Higgo and Rees (1986) also found that the initial sorption of plutonium onto clays was effective in removing most of the plutonium species that would be able to sorb onto the clay. When sorption to carbonate marine sediments was investigated, it was found that some desorption from the surface would also occur.

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This behavior was due to the presence of plutonium carbonate complexes on the sediment surfaces which were sorbed less strongly than plutonium dioxide complexes (Higgo and Rees 1986). In fact, the formation of plutonium complexes with organic carbon causes plutonium to remain in solution as a complex (NCRP 1984).

Plutonium can be taken up from various environmental media into plants and animals. The highest concentrations of plutonium in plants are found in the roots where plutonium is present as a surface-absorbed plutonium complex, a stabilized complex, or as a soluble plutonium complex (Garland et al. 1981). The concentration of plutonium in soil can be compared with the concentration in plants to determine what fraction present in soil reaches the plant. Soil to plant concentration ratios of 1×10^{-6} to 2.5×10^{-4} plutonium in wet vegetation/plutonium in dry soil have been calculated based on radioisotope experiments in plants grown in controlled environments. The stems and leaves have lower overall concentrations of plutonium than the roots, but higher concentrations of soluble plutonium. The seeds were found to have low concentrations of plutonium, which indicated that plutonium was not very mobile in plants (Cataldo et al. 1987). In studies on orange trees, Pinder et al. (1987) found that plutonium-238 was deposited on the leaf or soil surface, remained there, and that no measurable quantities were transferred to the fruits. Grain crops grown near the Savannah River Plant, Aiken, South Carolina, were found to contain higher concentrations of plutonium the closer to the facility they were grown. During harvesting, plutonium from soils or straw was resuspended and mixed with the crop. Plutonium in vegetable crops grown at Oak Ridge National Laboratory, Oak Ridge, Tennessee, contained higher plutonium concentrations in the foliage biomass than in the fruit. Peeling of potatoes and beets removed 99% of the residual plutonium (Adriano et al. 1980).

Plutonium transferred from soil or plants to grazing herbivores was predominantly associated with the animal's pelt and gastrointestinal tract (Hakonson and Nyhan 1980). Rodents studied near the Los Alamos and Trinity sites in New Mexico support this claim. Hakonson and Nyhan (1980) found no evidence of bioconcentration through the food chain from soil to plants to rodents. They concluded that soil was the source of plutonium in rodents. In contrast, a study by Sullivan et al. showed that rodents absorbed more plutonium-238 when it was incorporated into alfalfa (by growing it in soil containing plutonium) than when it was administered in the inorganic form (Sullivan et al. 1980). This study suggests that plutonium bound to organic compounds may have increased availability. However, the authors indicate that further study is needed.

Plutonium was found to bioaccumulate in aquatic organisms, primarily at the lower end of the food chain. The bioconcentration factors (i.e., the amount of the chemical found in the organism divided

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by the concentration in the surrounding water over the same time period) were 1,000 for mollusks and algae, 100 for crustacea, and 10 for fish (WHO 1983). Plutonium is concentrated in the bones of fish rather than in muscle tissues, as seen by whole fish to muscle tissue ratios of 2×10^6 to 5×10^4 or 40:1 (NCRP 1984).

5.3.2 Transformation and Degradation

Plutonium is formed and transmuted through radioactive decay. Three common types of radioactive processes involve the release of alpha or beta particles or gamma rays. Alpha decay results in the release of an alpha particle, which is a charged particle emitted from the nucleus of an atom having a mass and charge equal in magnitude to a helium nucleus (i.e., two protons and two neutrons). In alpha decay, the atomic mass of the nuclide is reduced by four and the atomic number by two. For example, plutonium-239 undergoes alpha decay to form uranium-235.

Beta particles are charged particles emitted from the nucleus of an atom with a mass and charge equal in magnitude to that of an electron. In beta decay reactions, as the electron is ejected, the number of protons in the resulting atom increases, changing the atomic number of the atom but not the mass. For example, plutonium-241 undergoes beta decay to form americium-241.

A gamma ray is short wavelength electromagnetic radiation emitted from the nucleus. Nuclei which have undergone transmutation by alpha or beta decay or by capture of a neutron often return to the ground state by emission of gamma radiation. Addition of a neutron changes the atomic mass or isotope number of the element but not the atomic number, as seen by the formation of plutonium-242 from plutonium-241.

The chemical transformation reactions plutonium undergoes in the environment are primarily oxidation and reduction reactions. There are five oxidation states found in the environment. These are plutonium(III), plutonium(IV), plutonium(V), plutonium(VI), and plutonium(VII). The last, plutonium(VII), is not commonly found and it is only found under very alkaline conditions. The dominant oxidation state of plutonium in the environment is plutonium(IV) (Wildung et al. 1987).

5.3.2.1 Air

Plutonium does not undergo transformation processes in the air beyond those related to radioactive decay. Radioactive decay will be important for the short-lived isotopes with half-lives less than the average residence time in the troposphere of approximately 60 days. For example, plutonium-237 has a half-life of 46 days and undergoes electron capture to form neptunium-237 which has a half-life of 2.1×10^6 years.

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(Nero 1979). Therefore, neptunium-237 may form in the stratosphere prior to deposition of plutonium-237 on the earth's surface as fallout.

5.3.2.2 Water

The important chemical transformation process in surface water is the oxidation or reduction of plutonium. In waters with low suspended solids, plutonium is generally found in oxidized forms, dissolved in the water. In waters with high suspended solids, plutonium is generally reduced and sorbed onto either suspended solids or sediments (Choppin and Morse 1987; Higgo and Rees 1986; Nelson et al. 1987).

Plutonium behaves differently than many other inorganic elements in that it can exist simultaneously in four oxidation states over a range of pH values. Under acidic conditions, the nature of the complexing ligands present in solution will influence the oxidation state of plutonium. The presence of fulvic acid (a naturally occurring organic acid) facilitates the reduction of plutonium(IV) to plutonium(III), especially below pH 3.1. The reduction of the higher oxidation states appears to be even less dependent on pH, especially below pH 6 (Bondietti et al. 1976).

5.3.2.3 Soil

Plutonium found in soils may undergo the same oxidation/reduction reactions described for surface waters in places where soil contacts water. In addition to oxidation/reduction reactions, plutonium can react with other ions in soil to form complexes. These complexes may then be absorbed by roots and move within plants; however, the relative uptake by plants is low. In plants, the complex can be degraded but the elemental plutonium will remain.

5.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

5.4.1 Air

Table 5-1 summarizes plutonium levels measured in air at nine different locations. Since 1945, when the first nuclear weapon test at Alamogordo, New Mexico, was conducted, approximately 360 kCi (1.3×10^{16} Bq) of plutonium-239, -240 have been released into the atmosphere from various sources. The accidental burn up of the SNAP-9 satellite added 17 kCi (6.3×10^{14} Bq) to the higher altitudes of the atmosphere (Perkins and Thomas 1980).

A 15-year study (1966 to 1980) in Palomares, Spain, reported levels of plutonium in air near the site of a crash between a United States military aircraft carrying four nuclear bombs, and a tanker aircraft following the cleanup of the contaminated area (Iranzo et al. 1987).

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TABLE 5-1. Plutonium Levels Detected in Air

Location	Quantity [pCi/m ³ (Bq/m ³)]	Study	Comments
Spain, weapon accident:			
Urban area	1.5×10^{-4} (5.5×10^{-6}) $^{239,240}\text{Pu}$	Iranzo et al. 1987	Average over 15 years
Near farm land	1.4×10^{-3} (5.2×10^{-5}) $^{239,240}\text{Pu}$		
Savannah River Plant:			
First year:	3.1×10^{-5} (1.2×10^{-6}) ^{238}Pu 1.5×10^{-5} (4.3×10^{-7}) $^{239,240}\text{Pu}$	Corey et al. 1982	Cumulative concentrations over one-year time period (not an average)
Second year:	1.15×10^{-3} (5.6×10^{-5}) ^{238}Pu 3.2×10^{-5} (1.2×10^{-6}) $^{239,240}\text{Pu}$		
Rocky Flats facility:			
1 m	2.2×10^{-3} (8.1×10^{-5}) $^{239,240}\text{Pu}$	Volchok et al. 1977	Average concentrations over 7 years
1 km	4.6×10^{-4} (1.7×10^{-5}) $^{239,240}\text{Pu}$		
2.5 km	7.0×10^{-5} (2.6×10^{-6}) $^{239,240}\text{Pu}$		
New York City	3.4×10^{-5} (1.3×10^{-6}) $^{239,240}\text{Pu}$	Volchok et al. 1977	Average concentrations over 7 years
New York City	3.0×10^{-5} (1.1×10^{-6}) ^{239}Pu 1.0×10^{-5} (3.7×10^{-7}) ^{238}Pu	Hardy 1973	Typical radioactivity concentrations in ground-level air (no further specification)

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Air sampling conducted continuously for 2 years (1975 to 1977) near the Savannah River Plant measured the average yearly concentrations of plutonium-238 and of plutonium-239, -240. The data for the first and second years of the study are presented in Table 5-1 (Corey et al. 1982). The data in Table 5-1 indicate that releases from the Savannah River Plant and levels detected in New York City (Hardy 1973) are on the same order of magnitude and are much lower than those observed in Palomares, Spain, following the cleanup of the weapon accident. Continuous air sampling in the vicinity of the Rocky Flats facility near Denver was initiated in 1970. The data in Table 5-1 are from locations 50 meters, 1 kilometer, and 2.5 km from the facility and are arithmetic means of data from sampling years 1970 to 1976. For purposes of comparison, sampling data for New York City for the same time interval are included. Data from all four locations indicated declining levels following 1971 (Volchok et al. 1977).

5.4.2 Water

Table 5-2 presents plutonium levels detected in several surface waters and groundwaters. The Pacific Ocean was sampled for plutonium and Northern Pacific concentrations were, on the average, greater than those detected in the Southern Pacific for both plutonium-239, -240 and plutonium-238 (Miyake and Sugimura 1976). The plutonium content of the particulate matter in three South Carolina estuarine systems was investigated by Hayes et al. (1976). The Neuse and Newport River estuaries received plutonium only through atmospheric fallout: the Savannah River estuary received effluent from the Savannah River Plant. Concentrations detected in the three estuaries are comparable. Raw water samples taken from three locations on the Savannah River were also found to contain levels comparable to the Savannah River estuary samples (Corey and Boni 1976). The estuarine and river concentrations were greater than the Pacific surface water samples, but were on the same order of magnitude as seawater samples taken from Trombay (Bombay, India) (Pillai and Mathew 1976). These results indicate that plutonium is found throughout the globe but that the highest concentrations of plutonium in water are found near source areas.

The groundwater at Enewetak Atoll and near the Idaho National Engineering Laboratory disposal well were found to contain plutonium-239, plutonium-240 and plutonium-238, respectively (Cleveland and Rees 1982; Noshkin et al. 1976). The isotope composition differs in the two areas (Table 5-2), and the levels detected in Idaho were, on average, lower than those detected at Enewetak Atoll. The range of groundwater concentrations at the Nevada Test Site was greater than the range detected in either of the other two groundwaters (Gonzalez 1988). Rainwater samples taken from Trombay (Bombay, India) in 1971 were reported to contain plutonium-239 concentrations greater than those

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TABLE 5-2. Plutonium Levels Detected in Water

Location	Quantity pCi/L (Bq/L)	Reference
North Pacific surface water	2.2×10^{-4} to 9.4×10^{-4} ^{238}Pu & $^{239,240}\text{Pu}$ (8.2×10^{-6} to 3.5×10^{-5})	Miyake and Sugimura 1976
South Pacific surface water	1.3×10^{-4} to 3.4×10^{-4} ^{238}Pu & $^{239,240}\text{Pu}$ (4.8×10^{-6} to 1.3×10^{-5})	
Enewetak, South Pacific: Groundwater	2.0×10^{-4} to 2.8×10^{-1} $^{239,240}\text{Pu}$ (7.4×10^{-6} to 1.0×10^{-2})	Noshkin et al. 1976
Idaho National Engineering Laboratory: Groundwater	1.1×10^{-2} to 7.8×10^{-2} ^{238}Pu (4.1×10^{-4} to 2.9×10^{-3})	Cleveland and Rees 1982
South Carolina: Estuarian waters	1.7×10^{-4} to 2.5×10^{-3} $^{239,240}\text{Pu}$ (6.3×10^{-6} to 9.4×10^{-5})	Hayes et al. 1976
River waters	4.3×10^{-4} to 2.3×10^{-3} $^{239,240}\text{Pu}$ (1.6×10^{-5} to 8.3×10^{-5})	Corey and Boni 1976
Trombay, India: Rainwater	8.2×10^{-2} (3.0×10^{-3}) ^{239}Pu 4×10^{-3} to 2×10^{-2} ^{239}Pu (1.5×10^{-4} to 7.4×10^{-4})	Pillai and Mathew 1976
New York City: Drinking water	8×10^{-5} to 6.1×10^{-4} $^{239,240}\text{Pu}$ (3.0×10^{-6} to 2.3×10^{-5})	Bogen et al. 1988
Nevada Test Site: Groundwater	4.2×10^{-2} to 2.6 ^{239}Pu (1.6×10^{-3} to 9.6×10^{-2})	Gonzalez 1988

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detected in seawater from the same area, as seen in Table 5-2 (Pillai and Mathew 1976).

Plutonium concentrations in the New York water supply measured between 1974 and 1979 showed a peak concentration of plutonium-239, -240 in the summer of 1974 which fell within the range of other surface waters analyzed (Table 5-2). The low concentration detected during the following autumn was below the concentrations detected in other waters (Bogen et al. 1988).

5.4.3 Soil

Average fallout levels in soils in the temperate United States are about 2,100,000 pCi/km² (7.8×10^4 Bq/km²) plutonium-239, -240 and 50,000 pCi/km² (1.9×10^3 Bq/km²) plutonium-238 (Hanson 1975). Mean deposition rates of stack-released plutonium-238 to soils around the Savannah River Plant range from 0.008 to 0.64 pCi/m²/day (3.0×10^{-4} to 2.4×10^{-2} Bq/m²/day). The range for plutonium-239, -240 was found to be 0.027 to 0.36 pCi/m²/day (1.0×10^{-3} to 1.33×10^{-2} Bq/m²/day). The lower concentrations were measured 9 km from the plant and the higher concentrations were measured 0.23 km from the plant (Corey et al. 1982).

Plutonium levels in soils at Rocky Flats, Colorado, were analyzed by Little and Whicker (1978). Plutonium concentrations in soil samples collected to a depth of 21 cm had plutonium concentrations ranging from 1,400 to 59,000 pCi/kg (52 to 2,200 Bq/kg). A recent study on particle size and radionuclide levels in Great Britain soils reported plutonium-238 levels detected at a range of 200 to 18,000 pCi/kg (7.4 to 676 Bq/kg) and plutonium-239, -240 levels detected at a range of 800 to 83,000 pCi/kg (29.6 to 3,070 Bq/kg) (Livens and Baxter 1988). Core samples of surface soil at the Maxey Flats facility, where radioactive wastes were buried, were reported to contain a mean concentration of 1.9×10^5 pCi/kg (67 Bq/kg) of plutonium-238 and 22,000 pCi/kg (8 Bq/kg) of plutonium-239 and plutonium-240 (NEA/OECD 1981).

Plutonium concentrations found in Lake Michigan sediments were reported to range from 35 to 250 pCi/kg dry sediment (9.5×10^{-22} to 6.8×10^{-21} Bq/kg) (Edgington et al. 1976). It was estimated in this report that radioactivity in the sediments was confined to the upper 6 cm of the sediments, and in many of the core samples, no radioactivity was detected below a depth of 3 cm.

5.4.4 Other Media

A 1972 study on plutonium levels in the diet reported concentrations of plutonium-239, -240 ranging from $<2 \times 10^{-7}$ pCi/g ($<7.4 \times 10^{-9}$ Bq/g) for canned fruit to 1.1×10^{-4} pCi/g (4.1×10^{-6} Bq/g) in shellfish (Bennett 1976b). Of the shellfish sampled in this report (clams and shrimp), clams showed eight times the levels of plutonium

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found in shrimp. Fish and shellfish sampled in the Windscale and Northeast areas of the Irish Sea were reported to contain between 2.0×10^{-4} pCi/g (7.4×10^{-6} Bq/g) (herring muscle) and 2 pCi/g (0.074 Bq/g) (soft parts of mussel) (Hetherington et al. 1976).

5.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

Daily ingestion of plutonium-239, -240 in food in Japan between 1978 and 1980 due to atmospheric fallout was estimated to be 0.0045 pCi/day (1.7×10^{-4} Bq/day) (Hisamatsu et al. 1987). In 1974, the mean intake of plutonium in New York City from all sources including tap water was reported to be 0.0044 pCi/day (1.6×10^{-4} Bq/day) (Bogen et al. 1988). This same value was reported for daily intakes of plutonium in Italy from 1975 to 1978 (Bennett 1976b). Ingested plutonium is poorly absorbed from the gastrointestinal tract, although the form of plutonium will influence absorption (ICRP 1982).

The isotope of plutonium inhaled will affect its behavior in the body. The bones and the liver are the primary organs for plutonium deposition following translocation in the body (ICRP 1982). However, McInroy et al. (1989) indicate that muscle tissue may also be a site of deposition. Plutonium-238 dioxide is more rapidly translocated from the lungs than plutonium-239 dioxide thereby causing more plutonium-238 to be concentrated in other body tissues (ICRP 1982).

Mean concentrations of plutonium-239, -240 in human tissues from autopsy specimens in Japan ranged from 0.00025 pCi/g (9.3×10^{-6} Bq/g) (cerebrum) to 0.0015 pCi/g (5.4×10^{-5} Bq/g) (gonads) fresh weight (Takizawa 1982).

Wrenn and Cohen (1977) reported plutonium-239 levels in tissues derived from 12 autopsy cases in New York City from 1973 to 1976. Average levels for lung, liver, vertebrae, and gonads were 0.00024 pCi/g of tissue (8.9×10^{-6} Bq/g), 0.0007 pCi/g (2.6×10^{-5} Bq/g), 0.00017 pCi/g (6.3×10^{-6} Bq/g), and 0.0004 pCi/g (1.5×10^{-5} Bq/g), respectively.

Tissue samples from autopsy cases of nonoccupationally exposed individuals from Great Britain showed median plutonium-239, -240 levels for ribs, vertebrae, femur, liver and lungs of 0.00016 pCi/g (5.9×10^{-6} Bq/g), 0.00012 pCi/g (4.4×10^{-6} Bq/g), 0.000095 pCi/g (3.5×10^{-6} Bq/g), 0.0007 pCi/g (2.6×10^{-5} Bq/g) and 0.000049 pCi/g (1.8×10^{-6} Bq/g), respectively. Comparable samples taken from autopsy cases from a region in Great Britain located near a plutonium processing plant had median concentrations of 0.00022 pCi/g (8.1×10^{-6} Bq/g), 0.00019 pCi/g (7.0×10^{-6} Bq/g), 0.00015 pCi/g (5.6×10^{-6} Bq/g), 0.00014 pCi/g (5.2×10^{-5} Bq/g) and 0.00018 pCi/g (6.7×10^{-6} Bq/g) for those tissues mentioned above (Poplewell et al. 1988).

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The estimated 50-year dose commitment from plutonium for people in the north temperate zone due to atmospheric tests conducted before 1973 is 0.2 mrad (0.002 mGy) to the bone lining cells (Eisenbud 1987). [The gray is an SI unit of absorbed dose and is equal to 0.01 rad.] The average annual dose equivalent from all background radiation to an individual residing in the United States is estimated to be 360 mrem (3.6 mSv) (NCRP 1987). [The sievert is an SI unit of dose equivalent and is equal to 0.01 rem.]

5.6 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

Individuals employed at facilities using plutonium or uranium have high exposure potentials. Voelz et al. (1985) studied workers who participated in the Manhattan Project to determine if they had been exposed to levels of plutonium which would result in adverse health effects. Examination of the study group showed that their health was as good, if not better, than the general population. Similar results were reported by Wilkinson et al. (1987) for individuals employed at a plutonium weapons facility. These authors found, however, that individuals with body burdens $\geq 2,000$ pCi (≥ 74 Bq) had a slightly higher mortality from all causes of death and from lymphopoietic neoplasms than that found in employees with body burdens $< 2,000$ pCi (< 74 Bq). (See Section 2.2.1.8 for a more complete discussion of this study.)

Cobb et al. (1982) obtained autopsy tissues from individuals who had lived in one of three areas around the Rocky Flats facility (449 decedents) and from individuals who had lived outside of these areas for use as control data. Total plutonium burden, as well as the ratio of plutonium-239, -240, were measured in lung and liver tissues from these individuals. Next of kin were interviewed to assure that none of the study population had been exposed to plutonium from sources other than fallout and/or environmental contamination from the Rocky Flats facility, and to obtain information on smoking history. Multiple regression analyses suggested that plutonium burden is related to age, sex, and smoking history, but showed no definitive relationship to residence near the Rocky Flats facility. The correlation of plutonium burden with smoking (measured in pack-years) indicated that smokers could be a population at risk for increased body burden. The authors of the study hypothesize that this may result from damage to the clearing mechanisms of the lungs, resulting in a decrease in the rate of natural elimination of particles.

Individuals living near facilities which utilize plutonium in the operations may have higher exposure potential due to regular releases through stack-emissions or waste water. In addition, atmospheric fallout to the soil can result in secondary releases due to dust generation while farming or due to uptake by crops and subsequent ingestion of contaminated crops (Corey et al. 1982).

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Individuals living in Palomares, Spain, were exposed to plutonium after the dispersal of the plutonium in two bombs released during the midair collision of two airplanes (Iranzo et al. 1987). Exposure via inhalation due to the resuspension of contaminated soil was studied for 15 years following the release. Those individuals living near cultivated lands with the highest contamination received a cumulative total of 52.3 mrem (5.2×10^{-1} mSv) from 1966 to 1980 while those in the urban area of Palomares, farther away from the source, received 5.4 mrem (5.4×10^{-2} mSv) (Iranzo et al. 1987).

Kathren et al. (1987) determined levels of plutonium-239 at autopsy in bones of an individual known to have had occupational exposure to plutonium. Values ranged from 1.9×10^{-4} to 1.14×10^{-2} pCi/g ash (7.0×10^{-6} to 5.0×10^{-5} Bq/g ash), with the highest value detected in the scapula. Kathren et al. (1988) found a greater percentage of plutonium-238 in the skeleton than plutonium-239.

Kawamura (1987) estimated the plutonium-239, -240 inhalation intake of visitors to Kiev after the Chernobyl accident to be 0.8 pCi/day (0.03 Bq/day) during peak fallout exposure.

5.7 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of plutonium is available. Where adequate information is not available, ATSDR, in conjunction with the NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of plutonium.

The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that, if met would reduce or eliminate the uncertainties of human health assessment. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

5.7.1 Identification of Data Needs

Physical and Chemical Properties. The physical and chemical properties of plutonium have been studied. The information is adequate to permit estimation of plutonium's environmental fate.

Production, Use, Release, and Disposal. The potential for human exposure to plutonium is great due to its ubiquitous presence in the environment, resulting from releases from production facilities and from

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weapons testing, and its radiological half-life. However, the level of exposure to plutonium may be small. The production and use of plutonium 238-243 are well documented. There is little information regarding the production of plutonium-237. The amounts of these plutonium isotopes produced for various applications have been documented; however, the most current information is from 1974. More recent data is needed in order to compare past and present production and to project future production. The majority of information on the production and use of plutonium is classified in the nation's defense program. Information on past major releases of plutonium from weapons testing and from the explosion of a navigational satellite is available. However, current information on releases from production facilities is unavailable and is needed in order to monitor populations that might be exposed. The disposal of plutonium prior to 1970 is documented, but again, more recent information regarding amounts being held for mandated disposal in the proposed high-level disposal facility is needed. Rules and regulations for the disposal of plutonium have been established and these are reported in Chapter 7.

Environmental Fate. The major transport processes involved in the environmental fate of plutonium, as it relates to potential human exposure, have been fairly well defined. These processes include transport in the atmosphere when adsorbed to particulate matter and dry or wet deposition on land and water. Information on environmental compartments, such as flux rates, and the mechanisms and rates of several processes involved in the biogeochemical cycling of plutonium are still undefined. The data available regarding uptake of plutonium by plants are limited. There is some information regarding the conversion of the oxidized forms of plutonium to reduced forms followed by uptake into plants. Information regarding the influence of inorganic complexes on transport and regarding the media-specific effects of pH on the oxidation states of plutonium would be useful in order to more fully understand transport processes. The persistence of plutonium isotopes is well documented. Transformation of plutonium is through radioactive decay or chemical oxidation/reduction reactions. These processes have been well characterized.

Bioavailability. Plutonium is known to be absorbed following inhalation exposure. Bioavailability following oral and dermal exposure is very low; however, plutonium can be absorbed from contaminated wounds. Bioassay data are available on absorption from contaminated air and water. However, information on the impact of the valence state of plutonium on absorption following oral exposure is ambivalent. Such information is needed in order to address the impact of chlorination of drinking water, which results in a change in the valence of plutonium from plutonium(IV) to plutonium(VI). Therefore, further testing is important to determine the relevance of this change in valence state. No data were located on absorption from ingestion of soil or plant

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material. Such information is needed in order to quantify the potential exposure by this route, particularly when children may ingest soil when playing near NPL sites.

Food Chain Bioaccumulation. Plutonium has been shown to bioconcentrate in aquatic organisms at the lower end of the food chain. However, data do not indicate that plutonium is bioconcentrated in plants, higher aquatic organisms, or animals. In addition, there is no indication that plutonium is biomagnified in terrestrial or aquatic food chains. No additional information on bioaccumulation appears to be necessary at this time.

Exposure Levels in Environmental Media. A number of studies have been performed throughout the years on the fallout associated with the testing of nuclear weapons. Information is available on levels in air, water, soil, plant materials, and foodstuffs. However, no recent data are available on levels in these media. In particular, information is very limited on levels in media associated with areas surrounding waste sites. Such information is needed in order to quantify the potential exposure via these sources. Data are not available on estimates of human intake via specific media. This information would be important in determining the impact of exposure through each of these media.

Exposure Levels in Humans. Plutonium is measurable in urine and in lung, liver, and bone tissues obtained from autopsy. It is plausible to expect that occupationally exposed populations are routinely biomonitored through urinalysis. However, such data are not made available and are needed to quantify exposure to these individuals. In addition, no information is available on biomonitoring of individuals around NPL sites where plutonium has been found or of the general public. This information is needed so that exposure to these populations may be quantified.

Exposure Registries. No exposure registries for persons environmentally exposed to plutonium were located. Plutonium is not currently one of the substances for which a subregistry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for subregistries to be established. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to the exposure to this substance.

5.7.2 On-going Studies

Long-term research studies on the environmental fate of plutonium have not been identified. However, with the Chernobyl accident, it is

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anticipated that new information regarding the transport and fate of materials released during the accident will become available.

N.P. Singh (University of Utah) is determining the concentration and accumulation of plutonium in different organs of the younger population of the United States, who were born after 1963. Another study includes determining the concentrations of plutonium-238, plutonium-239, and plutonium-240 in the liver, kidney, and bone of 30 people who lived in northern Utah.

C.R. Olsen (Oak Ridge National Laboratory), along with other researchers, is investigating whether radionuclides released from the Department of Energy Savannah River facilities might be useful environmental tracers. The study includes transport pathways, transfer rates, and geochemical fate of plutonium in the Savannah River estuary. R.E. Wildung at Pacific Northwest Laboratory is studying the influence of soil, soil microbial, and plant processes on behavior and cycling of cationic elements (including plutonium) in terrestrial environments.

The behavior of long-lived radionuclides in natural water (W.R. Penrose, Argonne National Laboratory) and the behavior of fallout-derived plutonium in estuarine sediments as a function of various environmental parameters (H.J. Simpson, Columbia University) have been under investigation. The study by Simpson includes determining factors which control the distribution of plutonium in the sediments of the Hudson River. Studies by Noshkin and Penrose include characterizing rates and mechanisms of various physical and chemical processes that control the behavior of such pollutants, and characterizing the importance of oxidation states and natural complexing agents on the sorption behavior of plutonium and other radionuclides.

Geochemistry of plutonium in the Gulf of Mexico is being studied by M.R. Scott (Texas A&M University). Plutonium isotopes will be measured in both oxic and anoxic sediments in the Gulf of Mexico and in suspended sediments from major rivers emptying into the Gulf.

G.R. Choppin (Florida State University) is investigating the synergistic reaction of actinide-TTA complexes with brown ether adducts in benzene solutions and interaction of plutonium and other transuranic elements with the components of marine sediments under different conditions. The interaction of plutonium-238 dioxide heat source with the marine environment is also under investigation by H.V. Weiss (Naval Coastal Systems Center).